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02/11/02

FORM PTO-1390 (Modified) (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER <b>32396</b>	
TRANSMITTAL LETTER TO THE UNITED STATES				U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR <b>10/049869</b> )	
DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371					
INTERNATIONAL APPLICATION NO. <b>PCT/EP99/05988</b>		INTERNATIONAL FILING DATE <b>12 August 1999</b>		PRIORITY DATE CLAIMED <b>12 August 1999</b>	
TITLE OF INVENTION <b>TERMOPLASTIC COMPOSITION</b>					
APPLICANT(S) FOR DO/EO/US <b>KERSTEN, Jean; LAMANT, Carole and SNAPS, Jean-Francois</b>					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.</li> <li>4. <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2))               <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau).</li> <li>b. <input type="checkbox"/> has been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).               <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is attached hereto.</li> <li>b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</li> </ol> </li> <li>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))               <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).</li> <li>b. <input type="checkbox"/> have been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).</li> <li>10. <input type="checkbox"/> An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).</li> <li>11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). <b>with Annexes:</b></li> <li>12. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210).</li> </ol>					
<b>Items 13 to 20 below concern document(s) or information included:</b>					
<ol style="list-style-type: none"> <li>13. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>15. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment.</li> <li>16. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li>17. <input type="checkbox"/> A substitute specification.</li> <li>18. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>19. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</li> <li>20. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</li> <li>21. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</li> <li>22. <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail <b>EV069013635US</b></li> <li>23. <input type="checkbox"/> Other items or information:</li> </ol>					

U.S. APPLICATION NO. (IF KNOWN) SEE 37 CFR <div style="font-size: 24pt; font-weight: bold; margin-top: 5px;">10/049869</div>		INTERNATIONAL APPLICATION NO. <div style="font-weight: bold; margin-top: 5px;">PCT/EP99/05988</div>		ATTORNEY'S DOCKET NUMBER <div style="font-weight: bold; margin-top: 5px;">32396</div>	
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24. The following fees are submitted:

**BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5) ) :**

<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO . . . . .	<b>\$1040.00</b>
<input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO . . . . .	<b>\$890.00</b>
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO . . . . .	<b>\$740.00</b>
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) . . . . .	<b>\$710.00</b>
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) . . . . .	<b>\$100.00</b>

**ENTER APPROPRIATE BASIC FEE AMOUNT =**

Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☒ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	44 - 20 =	24	x \$18.00	<b>\$432.00</b>
Independent claims	5 - 3 =	2	x \$84.00	<b>\$168.00</b>
Multiple Dependent Claims (check if applicable) <input type="checkbox"/>				<b>\$0.00</b>
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$1,620.00</b>
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.				<b>\$0.00</b>
<b>SUBTOTAL =</b>				<b>\$1,620.00</b>
Processing fee of <b>\$130.00</b> for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).				<b>\$0.00</b>
<b>TOTAL NATIONAL FEE =</b>				<b>\$1,620.00</b>
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/>				<b>\$0.00</b>
<b>TOTAL FEES ENCLOSED =</b>				<b>\$1,620.00</b>
				Amount to be: refunded \$
				charged \$

**CALCULATIONS PTO USE ONLY**

a. ☒ A check in the amount of   **\$1,620.00**   to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No.   **19-0522**  . A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

**Thomas H. Van Hoozer**  
**HOVEY WILLIAMS LLP**  
**2405 Grand Boulevard, Suite 400**  
**Kansas City, Missouri 64108**

**\*23589\***

PATENT TRADEMARK OFFICE

\_\_\_\_\_  
 SIGNATURE

**Thomas H. Van Hoozer**  
 \_\_\_\_\_  
 NAME

**32,761**  
 \_\_\_\_\_  
 REGISTRATION NUMBER

**February 11, 2002**  
 \_\_\_\_\_  
 DATE

JC11 Rec'd PCT/PTO 11 FEB 2002

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application of:

KERSTEN, Jean; LAMANT, Carole and  
SNAPS, Jean-Francois

U.S. National Phase Application  
of International Application Serial No. :  
PCT/EP99/05988

International Application Filing Date:  
August 12, 1999

THERMOPLASTIC COMPOSITION

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Assistant Commissioner of Patents  
Box PCT  
Washington, D.C. 20231

Sir:

**PRELIMINARY AMENDMENT**

Please enter the following preliminary amendment prior to examination of this application on the merits and computation of the fees:

**ABSTRACT**

Applicant submits herewith an abstract of the invention retyped on a separate page in conformance with U.S. practice. The abstract is taken from that appearing on the cover of the published international application.

**SPECIFICATION**

Please amend the specification as follows:

At page 1, line 2, please insert the heading:

-- Background of the Invention

1. Field of the Invention --.

At page 1, line 5, please delete "The prior art" and the underlining and substitute:

-- 2. Description of the Prior Art --.

At page 2, line 1, please delete "Brief description of the invention" and the underlining and substitute therefor:

-- Summary of the Invention --.

At page 10, line 17, please delete the underlining beneath the heading "Brief description of the drawings" so that it reads -- Brief description of the drawings -- .

At page 11, line 1, please delete the underlining beneath the heading "Examples of compositions according to the invention" so that it reads -- Examples of compositions according to the invention -- .

At page 11, line 4, please delete the underlining beneath the heading "Composition 1" so that it reads -- Composition 1 -- .

At page 11, line 27, please delete the underlining beneath the heading "Composition 2 to 5" so that it reads -- Composition 2 to 5 --.

At page 12, line 4, please delete the underlining beneath the heading "Composition 8 to 16" so that it reads -- Composition 8 to 16 -- .

At page 13, line 2, please delete the underlining beneath the heading "Compositions 17 to 24" so that it reads -- Compositions 17 to 24 -- .

At page 13, line 11, please delete the underlining beneath the heading "Compositions 25 to 33" so that it reads -- Compositions 25 to 33 -- .

At page 14, line 3, please delete the underlining beneath the heading "Compositions 34 to 42" so that it reads -- Compositions 34 to 42 -- .

At page 15, line 4, please delete the underlining beneath the heading "Compositions 43 to 58" so that it reads -- Compositions 43 to 58 -- .

At page 15, line 12, please delete the underlining beneath the heading "Examples of embodiments" so that it reads -- Examples of embodiments --.

At page 15, line 14, please delete the underlining beneath the heading "Example of stoppers" so that it reads -- Example of stoppers -- .

At page 15, line 16, please delete the underlining beneath the heading "Stopper n° 1" so that it reads -- Stopper n° 1 -- .

At page 17, line 2, please delete the underlining beneath the heading "Stopper n° 2" so that it reads -- Stopper n° 2 -- .

At page 17, line 11, please delete the underlining beneath the heading "Stopper n° 3" so that it reads -- Stopper n° 3 -- .

At page 17, line 23, please delete the underlining beneath the heading "Example of film" so that it reads -- Example of film -- .

At page 18, line 11, please delete the underlining beneath the heading "Example of tube" so that it reads -- Example of tube -- .

At page 19, line 3, please delete the underlining beneath the heading "CLAIMS" so that it reads -- CLAIMS --.

### CLAIMS

Please cancel claims 1 - 29 as presented in the Annexes to the International Preliminary Examination Report.

Please add the following new claims:

30. Thermoplastic composition comprising:
- a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin, in which the thermoplastic elastomer mixture further comprises:
- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and
  - a thermoplastic elastomer which is at least partially cross-linked, the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1.
31. The composition of claim 30, in which the not cross-linked SIS elastomer has a molecular weight comprised between 150,000 and 275,000.
32. The composition of claim 30, in which the not cross-linked SIS elastomer has a molecular weight comprised between 200,000 and 240,000.
33. The composition of claim 30, in which the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially not cross-linked and polyolefin resin is comprised between 1:5 and 1:1.

34. The composition of claim 30, in which the polyolefin resin is selected from the group consisting of polyethylene, polypropylene, and mixtures of polyethylene and polypropylene.

35. The composition of claim 30, in which the polyolefin resin and the at least partially cross-linked thermoplastic elastomer forms a premixture containing not cross-linked thermoplastic elastomer and partially cross-linked thermoplastic elastomer.

36. The composition of claim 30, in which the at least partially cross-linked thermoplastic elastomer has a cross-linking rate of more than 20%.

37. The composition of claim 30, in which the at least partially cross-linked thermoplastic elastomer has a cross-linking rate comprised between 25% and 75%.

38. The composition of claim 30, in which the composition comprises as the thermoplastic elastomer and polyolefin resin, a mixture of polyolefin resin and SIS elastomer, the said mixture containing at least 40% by weight SIS elastomer, whereby the said mixture contains at least 20% by weight at least partially cross-linked SIS elastomer.

39. The composition of claim 30, further comprising a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which has been partially cross-linked in the presence of polyolefin, the weight ratio of not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer which has been partly cross-linked in the presence of a polyolefin being comprised between 1:6 and 1:1.

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40. The composition of claim 30, further comprising a polyolefin resin, a partly cross-linked thermoplastic elastomer and a not cross-linked thermoplastic SIS elastomer, the weight content of partially cross-linked elastomer with respect to the weight of polyolefin resin, partially

cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 20% and 40%, while the weight content of not cross-linked thermoplastic SIS elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 15% and 50%.

41. The composition of claim 30, further comprising at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic SIS elastomer.

42. The composition of claim 41, in which the weight ratio of the not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer is lower than 1:2.

43. The composition of claim 30, further comprising at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic elastomer, the weight ratio of the not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer being lower than 1:10.

44. The composition of claim 30, in which the polyolefin resin and the thermoplastic elastomer(s) is a substantially homogeneous mixture of a substantially homogeneous premixture of a polyolefin and a partly cross-linked thermoplastic elastomer, with a not cross-linked thermoplastic SIS elastomer.

45. The composition of claim 30, in which a silane is used for the partial cross-linking of the thermoplastic elastomer.

46. The composition of claim 30, further comprising an amount of not cross-linked elastomer sufficient for ensuring a stability to a treatment selected from the group consisting



of heat treatment at a temperature of at least 121 °C of at least 100 minutes,  $\gamma$  irradiation of at least 20 KGray, and combinations thereof.

47. The composition of claim 30, further comprising at least an additive selected from the group consisting of dyes, pigments and mixtures thereof.

48. The composition of claim 30, which comprises less than 0.5% by weight halide salt.

49. The composition of claim 48, which comprises less than 0.2% by weight halide salt.

50. Sealing means for a container or vial, at least a part of the said sealing means being made of a composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin, in which the thermoplastic elastomer mixture comprises:

- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and
- a thermoplastic elastomer which is at least partially cross-linked, the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1.

51. The sealing means of claim 50, in which the not cross-linked SIS elastomer has a molecular weight comprised between 150,000 and 275,000.

52. The sealing means of claim 50, in which the not cross-linked SIS elastomer has a molecular weight comprised between 200,000 and 240,000.

53. The sealing means of claim 50, in which the polyolefin resin is selected from the group consisting of polyethylene, polypropylene, and mixtures of polyethylene and polypropylene.

54. The sealing means of claim 50, in which the polyolefin resin and the at least partially cross-linked thermoplastic elastomer forms a premixture containing not cross-linked thermoplastic elastomer and partially cross-linked thermoplastic elastomer.

55. The sealing means of claim 50, in which the composition further comprises as thermoplastic elastomer and polyolefin resin, a mixture of polyolefin resin and SIS elastomer, the said mixture containing at least 40% by weight SIS elastomer, whereby the said mixture contains at least 20% by weight at least partially cross-linked SIS elastomer.

56. The sealing means of claim 50, in which the composition further comprises a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which has been partially cross-linked in the presence of polyolefin, the weight ratio of not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer which has been partly cross-linked in the presence of a polyolefin being comprised between 1:6 and 1:1.

57. The sealing means of claim 50, in which the composition further comprises a polyolefin resin, a partly cross-linked thermoplastic elastomer and a not cross-linked thermoplastic SIS elastomer, the weight content of partially cross-linked elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 20% and 40%, while the weight content of not cross-linked thermoplastic SIS elastomer with respect to the weight of polyolefin resin, partially cross-linked

thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 15% and 50%.

58. The sealing means of claim 50, in which the composition further comprises at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic SIS elastomer.

59. The sealing means of claim 58, in which the weight ratio of the not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer is lower than 1:2.

60. The sealing means of claim 50, in which the polyolefin resin and the thermoplastic elastomer(s) is a substantially homogeneous mixture of a substantially homogeneous premixture of a polyolefin and a partly cross-linked thermoplastic elastomer, with a not cross-linked thermoplastic SIS elastomer.

61. The sealing means of claim 50, in which a silane is used for the partial cross-linking of the thermoplastic elastomer.

62. The sealing means of claim 50, in which the composition comprises an amount of not cross-linked elastomer sufficient for ensuring a stability to a treatment selected from the group consisting of heat treatment at a temperature of at least 121 °C during at least 100 minutes,  $\gamma$  irradiation of at least 20 KGray, and combinations thereof.

63. The sealing means of claim 50, in which the composition further comprises at least an additive selected from the group consisting of dyes, pigments and mixtures thereof.

64. The sealing means of claim 50, in which the composition comprises less than 0.5% by weight halide salt.

65. The sealing means of claim 64, in which the composition comprises less than 0.2% by weight halide salt.

66. In combination:

a pharmaceutical container or vial defining an inner volume and a surface;

sealing means for sealing said pharmaceutical container or vial, the said sealing means

comprising a body, at least a part of the body being made of a composition

comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer

and a polyolefin resin, in which the thermoplastic elastomer mixture comprises :

-- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and

-- a thermoplastic elastomer which is at least partially cross-linked, the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1,

the said body being associated to a layer contacting the surface of the vial or container when the sealing means closes the said container or vial.

67. The combination of claim 66, in which the polyolefin resin of the composition is selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-polypropylene and mixtures thereof, while the said layer is at least partly made of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-polypropylene and mixtures thereof.

68. An element selected from the group consisting of film, bag, cloth, protecting cloth, tube, cap, and cap for protecting a needle of a syringe, said element comprising at least a layer

made of a composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin, in which the thermoplastic elastomer mixture comprises:

- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and
- a thermoplastic elastomer which is at least partially cross-linked,

the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1.

69. The element of claim 68, in which the polyolefin resin of the composition is selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-polypropylene and mixtures thereof, while the element further comprises at least one layer consisting essentially of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixtures thereof.

70. The element of claim 68, which further comprises a second layer made of said composition.

71. Process for the manufacture of an article from a composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin in which the thermoplastic elastomer mixture comprises:

- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and
- a thermoplastic elastomer which is at least partially cross-linked,

the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and

5:1, in which the said composition is mixed at a temperature sufficient for the at least partial melting of the not cross-linked SIS elastomer, and in which the said composition is thereafter formed into the said articles.

72. The process of claim 71, in which the said composition contains at least one additive selected from the group consisting of dyes, pigments and mixtures thereof and having a melting point corresponding substantially to the melting point of the not cross-linked SIS elastomer, in which a control of the additive distribution in the formed articles is carried out, and in which the mixing step is controlled so as to reach a substantially homogeneous distribution of said additive in a part of the composition just before its formation into the said article.

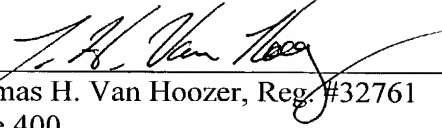
73. The process of claim 71, in which the said article is sterilized by a treatment selected from the group consisting of heating at a temperature of at least 121 °C during at least 100 minutes, irradiation with a  $\gamma$ -irradiation of at least 20 KGray, and combinations thereof.

### **REMARKS**

Applicant submits herewith a Preliminary Amendment for entry prior to computation of the fees and examination of the application on the merits. Applicant believes the amendment submitted herewith conforms the application to U.S. practice and it is believed that the amendment to the claims place them in allowable form. Early entry of the Notice of Allowance is courteously requested. Should this amendment necessitate any additional fees it may be charged to Deposit Account No. 19-0522.

Respectfully submitted,

HOVEY WILLIAMS LLP

By   
Thomas H. Van Hoozer, Reg. #32761  
Suite 400  
2405 Grand Boulevard  
Kansas City, Missouri 64108  
(816) 474-9050

(Docket No. 32396)

## ABSTRACT

Thermoplastic composition comprising a thermoplastic elastomer, a polyolefin resin, and possibly additive(s) and/or filler(s), characterized in that the thermoplastic elastomer is a mixture comprising a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which is at least partially cross-linked, the weight ratio not cross-linked thermoplastic SIS elastomer/thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1, preferably between 1:6 and 2.5:1.



Title: THERMOPLASTIC COMPOSITION  
Inventors: Kersten, Jean et al.  
U.S. National Phase Application corresponding to  
International Application PCT/EP99/05988  
Attorney Docket No.: 32396

**MARKED UP COPY OF PRELIMINARY AMENDMENT TO SHOW CHANGES MADE**

**SPECIFICATION**

Please amend the specification as follows:

- At page 1, line 2, please insert the heading:  
-- Background of the Invention  
1. Field of the Invention --.

- At page 1, line 5, please delete "The prior art" and the underlining and substitute:  
-- 2. Description of the Prior Art --.

At page 2, line 1, please delete "Brief description of the invention" and the underlining and substitute therefor:  
-- Summary of the Invention --.

At page 10, line 17, please delete the underlining beneath the heading "Brief description of the drawings" so that it reads -- Brief description of the drawings -- .

At page 11, line 1, please delete the underlining beneath the heading "Examples of compositions according to the invention" so that it reads -- Examples of compositions according to the invention -- .

At page 11, line 4, please delete the underlining beneath the heading "Composition 1" so that it reads -- Composition 1 -- .

At page 11, line 27, please delete the underlining beneath the heading "Composition 2 to 5" so that it reads -- Composition 2 to 5 --.

At page 12, line 4, please delete the underlining beneath the heading "Composition 8 to 16" so that it reads -- Composition 8 to 16 -- .

At page 13, line 2, please delete the underlining beneath the heading "Compositions 17 to 24" so that it reads -- Compositions 17 to 24 -- .

At page 13, line 11, please delete the underlining beneath the heading "Compositions 25 to 33" so that it reads -- Compositions 25 to 33 -- .

Title: THERMOPLASTIC COMPOSITION  
Inventors: Kersten, Jean et al.  
U.S. National Phase Application corresponding to  
International Application PCT/EP99/05988  
Attorney Docket No.: 32396  
Page2

At page 14, line 3, please delete the underlining beneath the heading "Compositions 34 to 42" so that it reads -- Compositions 34 to 42 -- .

At page 15, line 4, please delete the underlining beneath the heading "Compositions 43 to 58" so that it reads -- Compositions 43 to 58 -- .

At page 15, line 12, please delete the underlining beneath the heading "Examples of embodiments" so that it reads -- Examples of embodiments --.

At page 15, line 14, please delete the underlining beneath the heading "Example of stoppers" so that it reads -- Example of stoppers -- .

At page 15, line 16, please delete the underlining beneath the heading "Stopper n° 1" so that it reads -- Stopper n° 1 -- .

At page 17, line 2, please delete the underlining beneath the heading "Stopper n° 2" so that it reads -- Stopper n° 2 -- .

At page 17, line 11, please delete the underlining beneath the heading "Stopper n° 3" so that it reads -- Stopper n° 3 -- .

At page 17, line 23, please delete the underlining beneath the heading "Example of film" so that it reads -- Example of film -- .

At page 18, line 11, please delete the underlining beneath the heading "Example of tube" so that it reads -- Example of tube -- .

At page 19, line 3, please delete the underlining beneath the heading "CLAIMS" so that it reads -- CLAIMS --.

### CLAIMS

Please cancel claims 1 - 29 as presented in the Annexes to the International Preliminary Examination Report.

Please add the following new claims:

38. The composition of claim 30, in which the composition comprises as the thermoplastic elastomer and polyolefin resin, a mixture of polyolefin resin and SIS elastomer, the said mixture containing at least 40% by weight SIS elastomer, whereby the said mixture contains at least 20% by weight at least partially cross-linked SIS elastomer.

Title: THERMOPLASTIC COMPOSITION

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39. The composition of claim 30, further comprising a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which has been partially cross-linked in the presence of polyolefin, the weight ratio of not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer which has been partly cross-linked in the presence of a polyolefin being comprised between 1:6 and 1:1.

40. The composition of claim 30, further comprising a polyolefin resin, a partly cross-linked thermoplastic elastomer and a not cross-linked thermoplastic SIS elastomer, the weight content of partially cross-linked elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 20% and 40%, while the weight content of not cross-linked thermoplastic SIS elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 15% and 50%.

41. The composition of claim 30, further comprising at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic SIS elastomer.

42. The composition of claim 41, in which the weight ratio of the not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer is lower than 1:2.

43. The composition of claim 30, further comprising at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic elastomer, the weight ratio of the not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer being lower than 1:10.

44. The composition of claim 30 in which the polyolefin resin and the thermoplastic elastomer(s) is a substantially homogeneous mixture of a substantially homogeneous premixture of a polyolefin and a partly cross-linked thermoplastic elastomer, with a not cross-linked thermoplastic SIS elastomer.

45. The composition of claim 30, in which a silane is used for the partial cross-linking of the thermoplastic elastomer.

54. The sealing means of claim 50, in which the polyolefin resin and the at least partially cross-linked thermoplastic elastomer forms a premixture containing not cross-linked thermoplastic elastomer and partially cross-linked thermoplastic elastomer.

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55. The sealing means of claim 50, in which the composition further comprises as thermoplastic elastomer and polyolefin resin, a mixture of polyolefin resin and SIS elastomer, the said mixture containing at least 40% by weight SIS elastomer, whereby the said mixture contains at least 20% by weight at least partially cross-linked SIS elastomer.

56. The sealing means of claim 50, in which the composition further comprises a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which has been partially cross-linked in the presence of polyolefin, the weight ratio of not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer which has been partly cross-linked in the presence of a polyolefin being comprised between 1:6 and 1:1.

57. The sealing means of claim 50, in which the composition further comprises a polyolefin resin, a partly cross-linked thermoplastic elastomer and a not cross-linked thermoplastic SIS elastomer, the weight content of partially cross-linked elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 20% and 40%, while the weight content of not cross-linked thermoplastic SIS elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 15% and 50%.

58. The sealing means of claim 50, in which the composition further comprises at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic SIS elastomer.

59. The sealing means of claim 58, in which the weight ratio of the not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer is lower than 1:2.

60. The sealing means of claim 50, in which the polyolefin resin and the thermoplastic elastomer(s) is a substantially homogeneous mixture of a substantially homogeneous premixture of a polyolefin and a partly cross-linked thermoplastic elastomer, with a not cross-linked thermoplastic SIS elastomer.

61. The sealing means of claim 50, in which a silane is used for the partial cross-linking of the thermoplastic elastomer.

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62. The sealing means of claim 50, in which the composition comprises an amount of not cross-linked elastomer sufficient for ensuring a stability to a treatment selected from the group consisting of heat treatment at a temperature of at least 121 °C during at least 100 minutes,  $\gamma$  irradiation of at least 20 KGray, and combinations thereof.

63. The sealing means of claim 50, in which the composition further comprises at least an additive selected from the group consisting of dyes, pigments and mixtures thereof.

64. The sealing means of claim 50, in which the composition comprises less than 0.5% by weight halide salt.

65. The sealing means of claim 64, in which the composition comprises less than 0.2% by weight halide salt.

66. In combination:  
a pharmaceutical container or vial defining an inner volume and a surface;  
sealing means for sealing said pharmaceutical container or vial, the said sealing means comprising a body, at least a part of the body being made of a composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin, in which the thermoplastic elastomer mixture comprises :  
-- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and  
-- a thermoplastic elastomer which is at least partially cross-linked, the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1,  
the said body being associated to a layer contacting the surface of the vial or container when the sealing means closes the said container or vial.

67. The combination of claim 66, in which the polyolefin resin of the composition is selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-polypropylene and mixtures thereof, while the said layer is at least partly made of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-polypropylene and mixtures thereof.

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68. An element selected from the group consisting of film, bag, cloth, protecting cloth, tube, cap, and cap for protecting a needle of a syringe, said element comprising at least a layer made of a composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin, in which the thermoplastic elastomer mixture comprises:

- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and
  - a thermoplastic elastomer which is at least partially cross-linked,
- the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1.

69. The element of claim 68, in which the polyolefin resin of the composition is selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-polypropylene and mixtures thereof, while the element further comprises at least one layer consisting essentially of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixtures thereof.

70. The element of claim 68, which further comprises a second layer made of said composition.

71. Process for the manufacture of an article from a composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer and a polyolefin resin in which the thermoplastic elastomer mixture comprises:

- a not cross-linked thermoplastic SIS elastomer containing less than 20% by weight bound styrene, and
  - a thermoplastic elastomer which is at least partially cross-linked,
- the weight ratio of the not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1, in which the said composition is mixed at a temperature sufficient for the at least partial melting of the not cross-linked SIS elastomer, and in which the said composition is thereafter formed into the said articles.



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72. The process of claim 71, in which the said composition contains at least one additive selected from the group consisting of dyes, pigments and mixtures thereof and having a melting point corresponding substantially to the melting point of the not cross-linked SIS elastomer, in which a control of the additive distribution in the formed articles is carried out, and in which the mixing step is controlled so as to reach a substantially homogeneous distribution of said additive in a part of the composition just before its formation into the said article.

73. The process of claim 71, in which the said article is sterilized by a treatment selected from the group consisting of heating at a temperature of at least 121 °C during at least 100 minutes, irradiation with a  $\gamma$ -irradiation of at least 20 KGray, and combinations thereof.

2/Pt 2

1

Thermoplastic composition

The present invention relates to a thermoplastic elastomer composition.

5 The prior art

US-RE 32,028 discloses a thermoplastic mixture of partially cross-linked rubber with a polyolefin. This thermoplastic mixture is not suitable for the manufacture of sealing means for pharmaceutical vial, due to a lack of thermal stability and/or to a  
10 lack of irradiation stability, but essentially due to its easy fragmentation causing simple or multiple leaks after perforation with needles.

US 4,664,275 discloses sealing plug for pharmaceutical vial, said plug being made of a mixture containing partially cross-linked butyl rubber, a thermoplastic resin  
15 and an inorganic powder having a particle size lower than 150µm. The butyl rubber has a high flexural modulus and has to contain a high amount of inorganic solid particles so as to provide a good resistance to the passage of gases. The presence of such a high proportion of solid particles in the mixture requires a vigorous mixing step so to ensure a substantially homogeneous distribution of the  
20 particles in the mixture. Moreover, when puncturing such a plug with the needle of a syringe, a risk exists that solid particles are pulled away and fall into the pharmaceutical composition to be injected. Finally, the presence of such a high amount of solid particles restricts the possible re-use of the composition of the plugs, as said composition can only be re-used in the manufacture of articles in  
25 which the presence of such solid particles is authorized, and as the re-homogenization of a thermoplastic mixture comprising particles of butyl rubber mixed with solid particles is complex and/or difficult to reach. In addition, a high level of solid particles increases the risks of fragmentation and associated leakage post puncturing.

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Brief description of the invention

The present invention has for subject matter a thermoplastic composition which can be easily molded, injected or extruded without requiring the need of solid particles, a composition suitable for the manufacture of sealing plug ensuring a good gas  
5 sealing, an excellent closing of the passage due to the puncturing of the plug by means of a needle, and an excellent stability at a temperature of 121°C, as well as to  $\gamma$ -rays exposure. The composition according to the invention contains advantageously substantially thermoplastic components, where its manufacture, its  
10 use and its re-use is easy and not expensive.

The thermoplastic composition of the invention comprises a thermoplastic elastomer, a polyolefin resin, and possibly additive(s) and/or filler(s). The thermoplastic elastomer of the composition of the invention is a mixture  
15 comprising a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which is at least partially cross-linked, the weight ratio not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1, preferably between 1:6 and 2.5:1.

20 Not cross-linked thermoplastic SIS elastomer means a block copolymer styrene/isoprene/styrene or a mixture of block copolymer styrene/isoprene and block copolymer styrene/isoprene/styrene, said copolymer having an elasticity, a resilience, a melting point or a softening point suitable for the manufacture of closing plugs, etc. However, long-term performance of not cross-linked  
25 thermoplastic SIS elastomer is not warranted due to poor creep properties. Preferably the not cross-linked thermoplastic SIS elastomer contains less than about 20% by weight bound styrene, most preferably less than 18% by weight bound styrene, for example between 10 and 16% by weight bound styrene. The molecular weight is advantageously comprised between 150,000 and 275,000,  
30 preferably between 200,000 and 240,000. Preferably said not cross-linked thermoplastic SIS has a broad molecular weight distribution. According to an

embodiment, the molecular weight of the thermoplastic SIS is comprised between 0.9 and 1.1 the average molecular weight of the thermoplastic SIS ( the sum of the weight of the copolymer divided by the number of moles of copolymer).

- 5 The thermoplastic SIS elastomer has advantageously a very low halide salt content (due for example, to the use of halide salt, such as LiCl, for the copolymerisation process). For example the thermoplastic SIS contains less than 0.5% by weight halide salt with respect to the weight of not cross-linked thermoplastic SIS elastomer. Advantageously, the thermoplastic SIS contains less than 0.3% by weight, preferably less than 0.2% by weight, most preferably less than 0.1% by weight halide salt with respect to the weight of not cross-linked thermoplastic SIS elastomer.
- 10

- Therefore, the thermoplastic composition of the invention is substantially free of halide salt, for example contains less than 0.5% by weight halide salt, advantageously less than 0.3% by weight, preferably less than 0.2% by weight, most preferably less than 0.1% by weight halide salt.
- 15

- Thermoplastic elastomer which has been partially cross-linked means an elastomer with a melting point or a softening point corresponding to the temperature of manufacture of the articles, the said elastomer being preferably cross-linked in presence of a polyolefin resin. Said partially cross-linked thermoplastic elastomer is for example EPM elastomer, EPDM elastomer, SIS elastomer, or a mixture thereof, and is mixed with a polyolefin resin, such as polyethylene, polypropylene, copolymer of ethylene and propylene, or mixture thereof. By using an efficient amount of partially cross-linked thermoplastic elastomer (with respect to the amount of not cross-linked thermoplastic SIS elastomer) in the composition of the invention, the thermoplastic composition of the invention has improved long-term performance and other properties could be adjusted by varying the ratios.
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- 25

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Suitable polyethylenes which can be used in the composition of the invention are: ultra low density polyethylene (density of less than 0.91g/cm<sup>3</sup>), low density

polyethylene (density of less than 0.92 g/cm<sup>3</sup>), medium density polyethylene (0.926 to 0.94 g/cm<sup>3</sup>), high density polyethylene (more than 0.941 g/cm<sup>3</sup>), and mixture thereof.

- 5    Suitable polypropylenes are isotactic polypropylene, syndiotactic polypropylene, especially polypropylene with a high isotacticity, as well as copolymers with ethylene.

Copolymers of ethylene and propylene are for example amorphous copolymer  
10    (random distribution of the ethylene and propylene units), crystalline copolymer.

The polyolefin has advantageously a melt flow index (MFI) lower than 20, preferably comprised between 0.3 and 20 for a polypropylene and between 0.5 and 10 for a polyethylene.

15

The thermoplastic elastomer is preferably only partly cross-linked. The cross-linking rate of the said elastomer can be determined by measuring the gel content of the thermoplastic elastomer in cyclohexane (measured after immersion of the elastomer in cyclohexane during 48 hours at 73°F (23°C)). The gel content  
20    corresponds to the part of the elastomer which is insoluble in the cyclohexane. Said method is well-known in the art. A method for determining the cross-linking rate is disclosed in more details in ASTM D2765-90 (determination of gel content and swell ratio of cross-linked ethylene plastics).

- 25    The partial cross-linking of the elastomer is carried out by means of any known cross-linking agent. However, preferably, the cross-linking agent is a silane, for example a silane having hydrocarbon groups at least partly halogenated, a peroxide, organic peroxide, especially benzoyl peroxide and benzyl peroxide derivative. Other examples of possible peroxides are dicumyl peroxide, di-  
30    butylperoxide, 2,5-dimethyl-2,5-di-(tert-butylperoxy)hexane, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane-3, 1,3-bis(tert-butylperoxyisopropyl)benzene, 1,1-bis(tert-butylperoxy)-3,3,5-trimethylcyclohexane, n-butyl-4,4-bis(tert-

butylperoxy)valerate, p-chlorobenzoyl peroxide, 2,4-dichlorobenzoyl peroxide, tert-butylperoxybenzoate, tert-butylperoxyisopropyl carbonate, diacetyl peroxide, lauroyl peroxide, tert-butylcumyl peroxide, etc.

- 5 The partial cross-linking of the elastomer is preferably cross-linked in presence of a polyolefin resin, said cross-linking being advantageously carried out in a process in which the elastomer is partly cross-linked under conditions of high shear at a temperature above the melting point of the polyolefin component. The elastomer is thus simultaneously partly cross-linked and dispersed, for example as fine
- 10 particles, in the polyolefin. The partly cross-linking can be carried out in conventional mixing equipments, such as roll mills, bambury mixers, brabender mixers, continuous mixers, mixing extruders, etc. or in presence of water vapor for the silanes.
- 15 Preferably, in the composition according to the invention, the weight ratio not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked + polyolefin resin is comprised between 1:5 and 1:1, the polyolefin resin being advantageously selected from the group consisting of polyethylene, polypropylene, and mixture of polyethylene and polypropylene. Especially, the
- 20 polyolefin resin and the at least partially cross-linked thermoplastic elastomer forms a premixture containing not cross-linked thermoplastic elastomer and partially cross-linked thermoplastic elastomer distributed in the polyolefin.

According to an advantageous embodiment, the at least partially cross-linked

25 thermoplastic elastomer has a cross-linking rate of more than 20%, preferably comprised between 25% and 75 %. Said cross-linking rate corresponds to a gel content measured by the ASTM method ASTM D2765-90.

According to a particular embodiment, the composition of the invention comprises

30 as thermoplastic elastomer and polyolefin resin, a mixture of polyolefin resin and SIS elastomer, the said mixture containing at least 40% by weight SIS elastomer

According to a detail of a composition according to the invention, the polyolefin  
30 resin and the thermoplastic elastomer(s) is a substantially homogenous mixture of a  
substantially homogenous premixture of a polyolefin and a partly cross-linked  
thermoplastic elastomer, with a not cross-linked thermoplastic SIS elastomer. For

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The invention relates also to specific articles, a part of which is made of a composition according to the invention. Such articles can be manufactured by known techniques, such as molding, injection, extrusion, etc.

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Such an article is for example a sealing means for a container or vial (such as a plug or a stopper), at least a part of the said sealing means being made of a composition according to the invention.

- 10 Such a sealing means is for example intended for closing and sealing a pharmaceutical container or vial defining an inner volume, the said sealing means comprising a body, at least a part of which being made of a composition according to the invention. The said body can be associated to a layer contacting a surface of the vial or container when the sealing means closes the said container or vial.

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According to an advantageous embodiment, the sealing means is made at least partly of a composition according to the invention comprising a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof. When the said body is associated with a

20 contact or surface layer or provided with such a layer, the said layer is at least partly made of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof.

- 25 The composition according to the invention can be used for the manufacture of film, cloth, protecting cloth, such as finger protection, glove, etc. Preferably, the composition of the invention used comprises a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof. The film, cloth, etc has advantageously a multilayer structure,
- 30 at least one layer being made of the composition according to the invention, while at least another layer of the structure is essentially made of a polyolefin resin

selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof.

The film can be used for the manufacture of bags, injection bags, etc. For example, the film is folded so that parts of the film contact each other, parts of the film being  
5 thereafter welded together.

Tube, cap, cap for protecting a needle of a syringe, etc. can also be made at least partly of a composition in accordance to the invention. Advantageously, the tube, cap, etc comprises at least a layer made of a composition according to the  
10 invention.

According to a preferred embodiment, the tube comprises at least a first and a second layers made of a composition in accordance to the invention, the first layer having a not cross-linked SIS elastomer content lower than the not cross-linked  
15 SIS elastomer of the second layer. For example, the said first layer contacts and covers the second layer, i.e. the said second layer being directed towards the inner side of the tube with respect to the first layer.

The invention relates also to a process for the manufacture of an article from a composition according to the invention. In said process, the said composition is  
20 mixed at a temperature sufficient for the at least partial smelting of the not cross-linked SIS elastomer, and the said composition is thereafter transformed in the said articles.

25 Preferably, the said composition contains at least one dye or pigment having a melting point corresponding substantially to the melting point of the not cross-linked SIS elastomer. A control of the dye or pigment distribution in the transformed articles is carried out, and the mixing step is controlled so as to reach a substantially homogeneous distribution of the dye or pigment in a part of the  
30 composition just before its transformation in the said article. This quick control step is advantageous in order to determine whether an at least substantially constant quality of the manufactured articles can be reached.

After the manufacture of the articles, for example by injection or extrusion or molding, the manufactured articles are advantageously cooled by means of water, for example in a water bath.

The manufactured articles, preferably after their passage through a water bath, are  
5 advantageously sterilized at a temperature of 121°C during at least 100 minutes (for example during 180 minutes) and/or irradiated with a  $\gamma$ -irradiation of at least 20 KGray, for example 35 KGray.

Even if the composition of the invention ensures already excellent closing of the passage due to the puncturing of the plug by means of a needle, the composition  
10 can possibly contain additives, such as an isoprene derivative such as squalene, phytol, etc. (derivatives listed in column 4 of US 5,904,967, the content of which is incorporated herein by reference).

The composition of the invention contains however preferably less than 20% by weight of such isoprene derivative(s), most preferably less than 10% by weight, for  
15 example between 1 and 5% by weight.

#### Brief description of the drawings

Details and characteristics of the invention will appear from the following  
20 description in which reference is done to the attached drawings. In these drawings,  
- figure 1 is a cross section view of a stopper according to the invention ;  
- figure 2 is a cross section of another stopper according to the invention ;  
- figure 3 is a cross section of still another stopper according to the invention ;  
- figure 4 is a cross section of a film according to the invention ;  
25 - figure 5 is a partial perspective view of a bag according to the invention ; and  
- figure 6 is a cross section view of a tube according to the invention.

Examples of compositions according to the invention

Composition 1

5 In a tumbler, 60 parts by weight of a mixture polypropylene(PP)/EPDM (partly cross linked linear EPDM) sold under the name SANTOPRENE® 281-45 (sold by Advanced Elastomer Systems, USA, technical characteristics : free flowing pellets; tensile strength 3 MPa –ASTM D 412; elongation ultimate 300% -ASTM D 412;

10 density 0.97 g/cm<sup>3</sup>, hardness 45 Shore A –ASTM D 2240; processing temperature 175 to 230°C), 39 parts by weight of thermoplastic elastomer SIS ( a linear block copolymer sold by Shell Chemicals under the name KRATON D1161NS ®, technical characteristics : in the form of powder or pellets; density 0.92 g/cm<sup>3</sup> –

15 ISO 2781; Melt Flow Rate at 200°C, 5g : 12g/10minutes –ISO 1133 ; hardness, 30s : 30 Shore A – ISO 868; bound styrene content : about 15% by weight ; halide salt content : less than 0.2-0.3% by weight) and 1 part by weight of ultramarine blue (as master batch in polypropylene PP, melting point of about 160°C) have been mixed together.

20 The said mixture was used in a screw extruder so as to produce a continuous strip. The temperature of the extruder was about 180°C, while the screw was adapted so as to ensure a substantially homogeneous distribution of SIS elastomer in the Santoprene. The said homogeneous distribution is confirmed by the substantially uniform blue color of the strip during its extrusion.

25

Compositions 2 to 5

As for the composition 1, strips have been prepared by using SANTOPRENE – SIS

30 mixture, the composition of which is given in the following table.

Composition	2	3	4	5	6	7
SANTOPRENE % by weight	50	80	75	60	65	70
SIS % by weight	50	20	24	40	35	29
Ultramarine blue as master batch in PP % by weight			1			1

#### Compositions 8 to 16

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Strips have been prepared as for composition 1, except that SARLINK 3150N ® (a PP/radial cross linked EPDM sold by DSM, The Netherlands, technical features : specific gravity 0.94 g/cm<sup>3</sup>, Tensile strength 4.3 MPa – ISO 37; hardness, 5 seconds: 58°Shore A; Ultimate elongation 655%, melt temperature 185°C-220°C) has been used instead of SANTOPRENE 281. Details of the compositions are given in the following table.

10

composition	8	9	10	11	12	13	14	15	16
SARLINK % by weight	50	75	65	60	75	69	50	80	70
SIS % by weight	50	25	35	39	24	30	49	20	30
Ultramarine blue as master batch in PP % by weight				1	1	1	1		

Compositions 17 to 24

- Strips have been prepared as for composition 1, except that a mixture of SARLINK  
 5 3150N ® and SANTOPRENE 281 was used. Details of the compositions are given  
 in the following table.

Composition	17	18	19	20	21	22	23	24
SANTOPRENE % by weight	40	35	30	50	10	25	14	50
SARLINK % by weight	40	30	35	10	50	25	50	14
SIS	20	34	34	40	40	49	35	35
Ultramarine blue as master batch in PP % by weight		1	1			1	1	1

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Compositions 25 to 33

- In a mixer, a polyolefin has been mixed with SIS elastomer at 180°C. After  
 obtaining a substantially homogeneous mixture, benzoyl peroxide has been added  
 15 to the mixture as cross-linking agent in a quantity corresponding to about 5% of the  
 weight of the SIS elastomer and the partial cross linking was carried out at 180°C  
 under shearing in the extruder screw. About 80 % of the SIS was not cross-linked.  
 Possibly not cross-linked SIS elastomer was thereafter added.

- 20 The following table gives the composition of the thermoplastic elastomers  
 manufactured.

[illegible]

The said manufactured elastomer strips were submitted to a sterilizing treatment with water vapor (121°C) during 180 minutes or left in open air for one week.

#### Compositions 43 to 58

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These compositions have been prepared by adding to 100 parts by weight of one composition 1 to 16, y parts by weight of squalene. The following table gives the amount of squalene added.

Composition	100 parts by weight of composition n°	Y parts by weight of squalene
43	1	10
44	2	5
45	3	3
46	4	3
47	5	5
48	6	4
49	7	5
50	8	8
51	9	2
52	10	5
53	11	1
54	12	1
55	13	5
56	14	4
57	15	5
58	16	5

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#### Examples of embodiments

##### Example of stoppers

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##### Stopper n°1

The stopper of figure 1 is intended to close the opening of a neck of a pharmaceutical vial 100 (shown in dash line). Said stopper has a body 1 made of a composition according to the invention, the said body being provided on its face 10

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25 The elastomeric composition of the invention can be used for the manufacture of films or support or plates, such as mono layer film or multi layer film. Figure 4 shows in cross section the structure of a multi layer film, said film having a central portion 50 made of the composition according to the invention, and two outer layers 51,52, preferably made of a polyolefin, such as polyethylene, polypropylene, 30 copolymer of ethylene and propylene. The total thickness of the film is for example 50  $\mu\text{m}$  to 10 mm, especially between 100  $\mu\text{m}$  and 2 mm. The film can be produced by co extrusion and can be subjected to a stretching (mono axial or

biaxial). The film can be used for the manufacture of stoppers, plugs, etc by thermoforming.

5 The film can also be used for the preparation of bags. The film is folded so that parts of the film contact each other. Thereafter, by means of a welding machine, a welding 53 between parts of the inner film 51 is formed along the edge of the bag.

The film can also be used for the manufacture of gloves, protecting fingers, strips to be placed on the skin of a patient where an injection has to be made.

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#### Example of tube

The composition of the invention can be used for the manufacture of tube 60. Such a tube is advantageously a multi layer tube, which can be manufactured by co  
15 extrusion.

The tube has for example an inner compatible layer 61, for example a layer containing a few SIS elastomer, for example a polyolefin layer containing less than 10% SIS, a central layer 62 made of a composition according to the invention, and  
20 an outer polyolefin layer 63 containing no or a few SIS for giving a good abrasion resistance. The central layer 62 acts as a layer against leaks, even when the tube is punctured for example by means of a needle of syringe. Such a tube can be used for the manufacture of artificial veins, injection tube, for example a tube extending between a bag or baxter and a patient, whereby a pharmaceutical can be injected in  
25 the tube without creating a leakage, so that said pharmaceutical is injected through the injection site of the bag or baxter. (2)

*such as bags sold under the trademark*

AMENDED SHEET

## Amended claims

1. Thermoplastic composition comprising a thermoplastic elastomer mixture containing at least a styrenic elastomer, a polyolefin resin, and possibly additives(s) and/or filler(s), in which the thermoplastic elastomer mixture comprises :
  - a not cross-linked thermoplastic SIS elastomer contains less than 20% by weight bound styrene, advantageously less than 18% by weight bound styrene, preferably from 10 to 16% by weight bound styrene, and
  - a thermoplastic elastomer which is at least partially cross-linked, the weight ratio not cross-linked thermoplastic SIS elastomer/thermoplastic elastomer at least partially cross-linked being comprised between 1 :10 and 5 :1, preferably between 1 :6 and 2.5 :1.
2. The composition of claim 1, characterized in that the not crosslinked SIS elastomer has a molecular weight comprised between 150,000 and 275,000, advantageously between 200,000 and 240,000.
3. The composition of claim 1, characterized in that the at least partially cross-linked thermoplastic elastomer is a thermoplastic elastomer at least partially cross-linked in presence of a polyolefin resin.
4. The composition of claim 1 or 3, characterized in that the weight ratio not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer at least partially cross-linked + polyolefin is comprised between 1:5 and 1:1.
5. The composition of claim 1, characterized in that the polyolefin resin is selected in the group consisting of polyethylene, polypropylene, and mixture of polyethylene and polypropylene.
6. The composition of claim 1, characterized in that the polyolefin resin and the at least partially cross-linked thermoplastic elastomer forms a premixture containing not cross-linked thermoplastic elastomer and partially cross-linked thermoplastic elastomer.
7. The composition of claim 1, characterized in that the at least partially cross-linked thermoplastic elastomer has a cross-linking rate of more than 20%, preferably comprised between 25% and 75 %.

AMENDED SHEET

87. The composition of claim 1, characterized in that it comprises as thermoplastic elastomer and polyolefin resin, a mixture of polyolefin resin and SIS elastomer, the said mixture containing at least 40% by weight SIS elastomer, whereby the said mixture contains at least 20% by weight at least partially cross-linked SIS elastomer.
98. The composition of claim 1, characterized in that it comprises a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which has been partly cross-linked in presence of a polyolefin, the weight ratio not cross-linked thermoplastic SIS elastomer / thermoplastic elastomer which has been partly cross-linked in presence of a polyolefin being comprised between 1:6 and 1:1, advantageously 1:4 and 7:10, preferably between 1:3 and 1:2.
109. The composition of claim 1, characterized in that it comprises a polyolefin resin, a partially cross-linked thermoplastic elastomer and a not cross-linked thermoplastic SIS elastomer, the weight content of partially cross-linked elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 20 and 40%, while the weight content of not cross-linked thermoplastic SIS elastomer with respect to the weight of polyolefin resin, partially cross-linked thermoplastic elastomer and not cross-linked thermoplastic SIS elastomer being comprised between 15 and 50%.
110. The composition of claim 1, characterized in that it comprises at least 20% by weight of a not cross-linked thermoplastic elastomer different from the not cross-linked thermoplastic SIS elastomer.
111. The composition of claim 10, characterized in that the weight ratio not cross-linked thermoplastic elastomer different from the thermoplastic SIS elastomer / not cross-linked SIS thermoplastic elastomer is lower than 1:2, preferably lower than 1:10.

AMENDED SHEET

<sup>13</sup>  
12. The composition of claim 1, characterized in that the polyolefin resin and the thermoplastic elastomer(s) is a substantially homogenous mixture of a substantially homogenous premixture of a polyolefin and a partly cross-linked thermoplastic elastomer, with a not cross-linked thermoplastic SIS elastomer.

<sup>14</sup>  
13. The composition of claim 1, characterized in that a silane is used for the partial cross-linking of the thermoplastic elastomer.

10 <sup>15</sup>  
14. The composition of claim 1, characterized in that it comprises an amount of not cross-linked elastomer sufficient for ensuring a thermal stability at 121°C of at least 100 minutes and/or a  $\gamma$  irradiation stability of at least 20 KGray.

<sup>16</sup>  
15. The composition of claim 1, characterized in that it comprises at least a dye or a pigment.

<sup>17</sup>  
16. The composition of claim 1, characterized in that it comprises less than 0.5% by weight halide salt.

20 <sup>18</sup>  
17. The composition of claim <sup>17</sup>16, characterized in that it comprises less than 0.3% by weight, preferably less than 0.2% by weight, most preferably less than 0.1% by weight halide salt.

<sup>19</sup>  
18. Sealing means for a container or vial, at least a part of the said sealing means being made of a composition according to one of the preceding claims.

20 <sup>20</sup>  
19. The sealing means according to claim <sup>18</sup>18, for sealing a pharmaceutical container or vial defining an inner volume, the said sealing means comprising a body, at least a part of which being made of a composition according to one of the claims 1 to <sup>18</sup>18, the said body being associated to a layer contacting a surface of the vial or container when the sealing means closes the said container or vial.

- <sup>21</sup>  
20. The sealing means according to claim <sup>19</sup>~~18~~, characterized in that the polyolefin resin of the composition in accordance to one of the claims 1 to 17 is selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof, while the said layer is at least partly made of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof.
- <sup>22</sup>  
10 <sup>21</sup>  
21. Film, cloth, protecting cloth comprising at least a layer made of a composition in accordance to one of the claims 1 to <sup>19</sup>~~17~~.
- <sup>23</sup>  
15 <sup>22</sup>  
22. The film, cloth or protecting cloth of claim <sup>21</sup>~~20~~, characterized in that the polyolefin resin of the composition in accordance to one of the claims 1 to 17 is selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof, while the film, cloth or protecting cloth comprises at least one layer essentially made of a polyolefin resin selected from the group consisting of polyethylene, polypropylene, copolymer of ethylene-propylene and mixture thereof.
- <sup>24</sup>  
20 <sup>23</sup>  
23. Tube or cap, cap for protecting a needle of a syringe, the said tube or cap comprising at least one layer made of a composition in accordance to one of the claims 1 to <sup>19</sup>~~17~~.
- <sup>25</sup>  
25 <sup>24</sup>  
24. Tube in accordance to claim <sup>23</sup>~~22~~, characterized in that it comprises at least a first and a second layers made of a composition in accordance to one of the claims 1 to <sup>19</sup>~~17~~, a first layer having a not cross-linked SIS elastomer content lower than the not cross-linked SIS elastomer of the second layer.
- <sup>26</sup>  
30 <sup>25</sup>  
25. Bag made of a film according to claim <sup>21</sup>~~20~~ or <sup>23</sup>~~22~~.
- <sup>27</sup>  
26. Process for the manufacture of an article from a composition according to one of the claims 1 to <sup>19</sup>~~17~~, in which the said composition is mixed at a temperature

sufficient for the at least partial smelting of the not cross-linked SIS elastomer, and in which the ~~said~~ composition is thereafter <sup>shaped</sup> ~~transformed~~ in the said articles.

<sup>28</sup>  
27. The process of claim <sup>27</sup> 26, in which the said composition contains at least one  
5 dye or pigment having a melting point corresponding substantially to the melting  
point of the not cross-linked SIS elastomer, in which a control of the dye or  
pigment distribution in the transformed articles is carried out, and in which the  
mixing step is controlled so as to reach a substantially homogeneous distribution of  
the dye or pigment in a part of the composition just before <sup>the shaping of</sup> ~~its transformation~~ in the  
10 said article.

<sup>28</sup>  
28. The process of claim <sup>28</sup> 27, in which the said article is sterilized at a temperature  
of 121°C during at least 100 minutes and /or irradiated with a  $\gamma$ -irradiation of at  
least 20 KGray.

15



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*For two-letter codes and other abbreviations, refer to the "Guid-  
ance Notes on Codes and Abbreviations" appearing at the begin-  
ning of each regular issue of the PCT Gazette.*



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(54) Title: THERMOPLASTIC COMPOSITION

(57) Abstract: Thermoplastic composition comprising a thermoplastic elastomer, a polyolefin resin, and possibly additive(s) and/or filler(s), characterized in that the thermoplastic elastomer is a mixture comprising a not cross-linked thermoplastic SIS elastomer and a thermoplastic elastomer which is at least partially cross-linked, the weight ratio not cross-linked thermoplastic SIS elastomer/thermoplastic elastomer at least partially cross-linked being comprised between 1:10 and 5:1, preferably between 1:6 and 2.5:1.

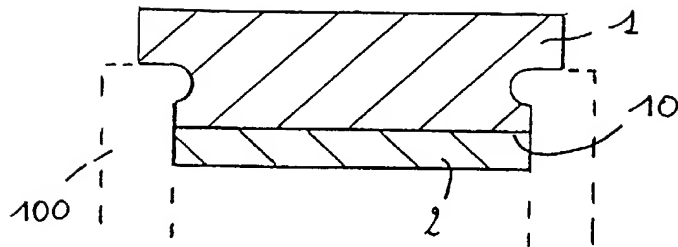


FIG 1

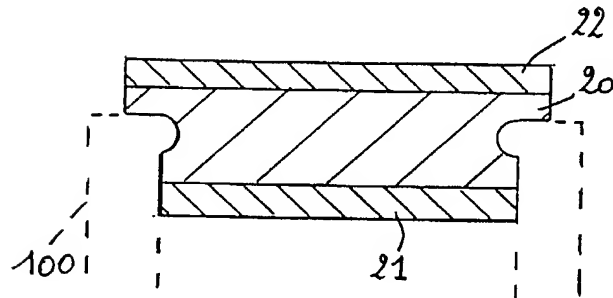


FIG 2

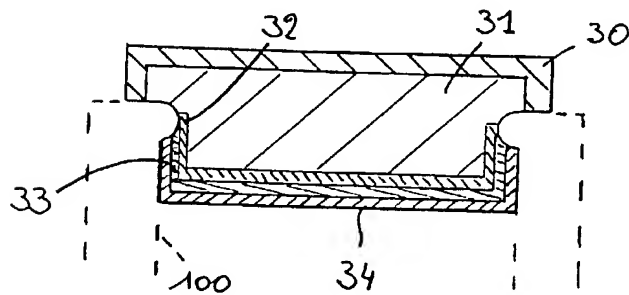


FIG 3

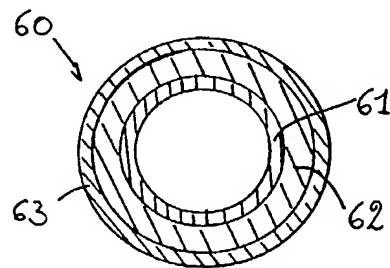
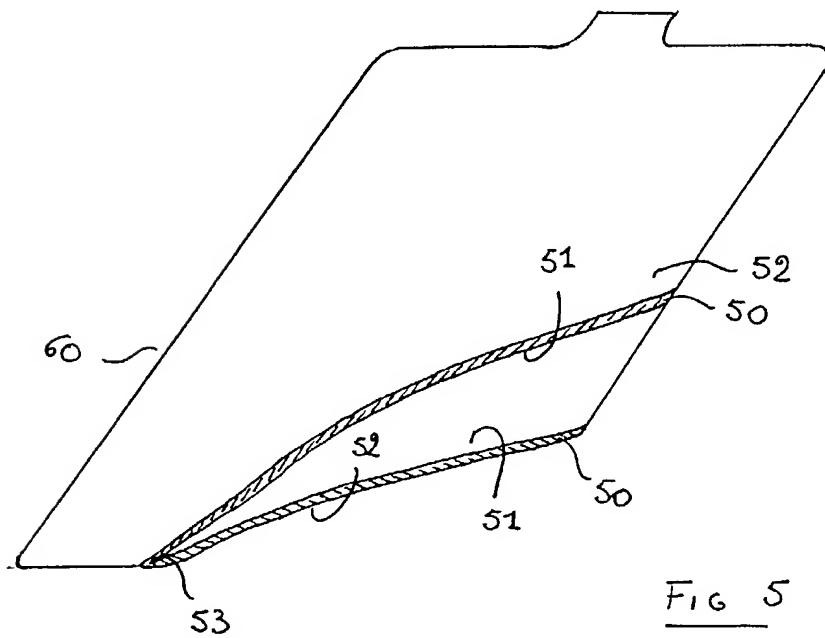
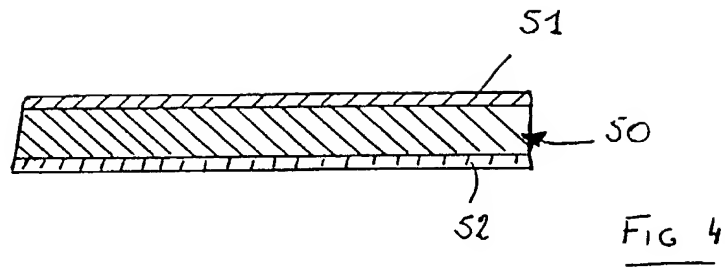


FIG 6



**COMBINED DECLARATION AND POWER OF ATTORNEY**  
 (Original, Design, National Stage of PCT  
 or CIP Application)

**ATTORNEY'S DOCKET NO.**  
 32396

As a below named inventor I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**THERMOPLASTIC COMPOSITION**

the specification of which: (complete (a), (b) or (c) for type of application)

**REGULAR OR DESIGN APPLICATION**

 (a) ☒ is attached hereto.

 (b) ☐ was filed on \_\_\_\_\_ as Application Serial No. \_\_\_\_\_  
 and was amended on \_\_\_\_\_ (if applicable).

**PCT FILED APPLICATION ENTERING NATIONAL PHASE**

 (c) ☒ was described and claimed in International Application No. PCT/EP99/05988 filed 12 AUG 1999  
 and as amended on \_\_\_\_\_ (if any).

**ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR**

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56(a).

☐ In compliance with this duty there is attached an information disclosure statement. 37 CFR 1.97.

**PRIORITY CLAIM**

I hereby claim foreign priority benefits under Title 35, United States Code, § 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed: (complete (d) or (e))

 (d) ☐ no such applications have been filed.

 (e) ☐ such applications have been filed as follows

**EARLIEST FOREIGN APPLICATION(S), IF ANY FILED WITHIN 12 MONTHS PRIOR TO SAID APPLICATION**

Country	Application No.	Date of Filing	Date of Issue	Priority Claimed
				<input type="checkbox"/> YES <input type="checkbox"/> NO
				<input type="checkbox"/> YES <input type="checkbox"/> NO
				<input type="checkbox"/> YES <input type="checkbox"/> NO

**ALL FOREIGN APPLICATION(S), IF ANY FILED MORE THAN 12 MONTHS PRIOR TO SAID APPLICATION**

Country	Application No.	Date of Filing	Date of Issue	Priority Claimed

PROVISIONAL

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States application(s) listed below:

Application Serial No.	Filing Date	Status (patented, pending, abandoned)
------------------------	-------------	---------------------------------------

CONTINUATION-IN-PART

(Complete This Part Only If This Is A Continuation-In-Part Application)

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56(a), which became available between the filing date of the prior application and the national or PCT international filing date of the continuation-in-part application:

Application Serial No.	Filing Date	Status (patented, pending, abandoned)
------------------------	-------------	---------------------------------------

POWER OF ATTORNEY

As a named inventor, I hereby appoint the following attorney(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

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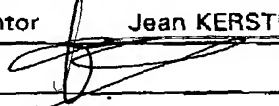
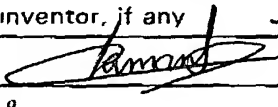
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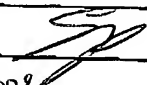
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(816) 474-9050

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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